

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION

OF: WITTELER ET AL.

SERIAL NO. 10/070,758

FILED: JULY 11, 2003

FOR: PROCESS FOR PREPARING POLYVINYLPIRROLIDONE-IODINE IN
AQUEOUS SOLUTIONHonorable Commissioner
for Patents
P.O. Box 1450
Alexandria, VA 22313-1450D E C L A R A T I O N

I, Dr. Rainer Dobrawa, Dr. rer. nat., a citizen of the Federal Republic of Germany and residing at Stamitzstrasse 18, 68167 Mannheim, Germany, hereby declare as follows:

I am fully trained chemist, having studied at the University of Ulm, Germany, from 1995 to 2000, from which I received my Diploma in Chemistry;

From 2000 to 2004 I furthered my studies at the Institute of Organic Chemistry of the University of Würzburg, Germany, and I was awarded my doctor's degree by the said university in 2004;

I joined BASF Aktiengesellschaft of 67056 Ludwigshafen, Germany, in 2004, and have since been working in the field of polymer research, and am therefore fully conversant with the technical field to which the invention disclosed and claimed in application Serial No. 10/070,758 belongs.

I have studied the record of application Serial No. 10/070,758, and particularly the Office action mailed on January 4, 2006, and the

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prior art applied by the Examiner, in particular the teaching of Denzinger et al. (US 4,402,937).

It is my understanding that the Examiner contends that the particular features of the process according to the claims of application Serial No. 10/070,758, are already known from Denzinger et al..

I cannot share the Examiner's position as set forth in the Office action of January 4, 2006, for the following reasons. The teaching Denzinger et al. relates to a process for the preparation of PVP-iodine by reacting elemental iodine and PVP in aqueous solution. In col. 3 indicated lines 37 to 41 of US 4,402,937 it is stated that PVP having a K value from 8 to 50 can be employed. In col. 4 indicated lines 34 to 37 of US 4,402,937 it is stated that the PVP can be employed in a concentration from 10 to 60 % by weight.

The Examiner's position that Denzinger et al.'s disclosure allows for a combination of any of the K values and any of the PVP concentrations which fall within the ranges taught by Denzinger et al. is not deemed to be well taken. It is well known in the polymer art that, at the same concentration, a solution of a PVP having a high K value is more viscous than a solution of a PVP having a low K value. Accordingly, when solutions of identical viscosity are prepared from PVPs having different K values the concentration of the solution containing the PVP with lower K value is higher than the concentration of the solution containing the PVP with the higher K value. This general background knowledge is reflected by the general teaching of Denzinger et al. in col. 4 indicated lines 34 to 37 where it is stated that the higher concentrations apply to PVP having a low K value, and vice versa. This general teaching is confirmed by the working examples of Denzinger et al.. In example 3 a PVP having a K value of 12.5 is employed at concentration of 50 % by weight, while in examples 2, 4, 5a-c PVPs having K-values of 16.6 or 17 are employed at concentration of 40 % by weight and in example 1 a PVP having a K value of 31.5 is employed at a concentration of 30 % by weight.

The process according to the claims of application Serial No. 10/070,758 requires that an aqueous PVP solution is employed which has PVP concentration c that is larger than value resulting from the calculation $100 \times [0.1 + 8:(K + 5)]$. Denzinger et al. do not teach the utilization of PVP solutions which meet this requirement. In fact, each

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of the working examples of *Denzinger et al.* utilize PVP solutions having a PVP concentration that is lower than the value obtained from the calculation of claim 1. In Example 1, *Denzinger et al.* employ a PVP having K value of 31.5 in a concentration of 30 % by weight. However, the minimum concentration required according to the calculation is 31.9 % by weight. In Examples 2 and 5a to 5c, *Denzinger et al.* employ a PVP having K value of 16.6 in a concentration of 40 % by weight. However, the minimum concentration required according to the calculation is 47 % by weight. In Example 3, *Denzinger et al.* employ a PVP having K value of 12.5 in a concentration of 50 % by weight. However, the minimum concentration required according to the calculation is 55.7 % by weight. In Example 4, *Denzinger et al.* employ a PVP having K value of 17 in a concentration of 40 % by weight. However, the minimum concentration required according to the calculation is 46.4 % by weight.

The inventors of application Serial No. 10/070,758 found out that the reaction taught by *Denzinger et al.* has several drawbacks. When performing the reaction of *Denzinger et al.* with a polyvinylpyrrolidone having a K-value of 30, mixing problems occur and practically insoluble sediments form as a result of mixing problems caused by the high viscosity of the PVP-solution. The sediment is presumably a complex mixture of PVP, iodine and probably overiodinated PVP. Moreover, the reaction is rather slow. When the reaction of *Denzinger et al.* is performed with a polyvinylpyrrolidone having a K-value of 12 strong sublimation of Iodine occurs, which leads to a clogging of the equipment. In both cases, the available iodine of the obtained PVP-iodine is not satisfactory. Therefore, the reaction of *Denzinger et al.* is of limited applicability for the production of PVP-iodine. The inventors surprisingly found out that this drawbacks can be overcome by employing aqueous PVP solutions which fulfill the requirements of the calculation of claim of application Serial No. 10/070,758, i.e. by employing an aqueous PVP solution having a concentration which is larger than the value obtained from said calculation.

These findings are supported by the following comparative examples, which were run under my supervision.

The reaction of the aqueous PVP solution and iodine was performed in a 2-Liter glass reactor equipped with a crossbeam agitator at 400 rpm, a condenser and an internal temperature control. The reactor

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was heated by an external heating bath. If not stated otherwise, 330 g of PVP were charged as an aqueous solution into the reactor. The concentration is given in the following table. To this solution 3,72 g of formic acid were added. The solution was heated to 85°C internal temperature. Then 70,00 g of elemental iodine were added in 10 portions, with 5 min between each addition of iodine. After iodine addition was completed, the solution was stirred at 85°C until the reaction was complete. For these experiments, the reaction is deemed to be completed when in the solution solid components can no longer be observed, which indicates that all solid iodine is dissolved and has reacted. The solution was subsequently cooled to room temperature. The resulting PVP-iodine solution was decanted and the degree of formation of solid lumps in the reactor as well as incrustations in the condenser is assessed. Iodine loss was determined according to procedure given on page 11 of application Serial No. 10/070,758 and available iodine was determined by titration with thiosulfate. The results are given in the following table:

Example	c ¹⁾ [%bw]	K value	c _{min} ²⁾ [%bw]	Obser- vations ³⁾	Available Iodine [%] ⁴⁾	Iodine Loss [%] ⁴⁾	reaction time
1	33	30	32.9	+	10.08	3.92	6 h
2	43	30	32.9	+	10.03	2.33	6 h
Comp.1	23	30	32.9	-	7.42	3.90	>10 h
3	33	60	22,3	+	9.75	4.33	>10 h
Comp.2	33	12	57.1	o	9.16	18.93	6 h

- 1) PVP concentration of the utilized PVP solution in % by weight
- 2) minimum concentration calculated according

$$c_{min} = 100 \times [0.1 + 8 : (K + 5)]$$
- 3) + no sedimentation, homogeneous mixing, no detectable solids
 o few solids, acceptable mixing, strong iodine sublimation
 - strong sedimentation, bad mixing, large amount of solid residue, iodine sublimation into the condenser
- 4) % by weight


The reactions according to examples 1, 2, and 3 were good to handle, and neither sedimentation nor marked sublimation of iodine was observed. For each example the available iodine was above 9.5% and the

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iodine loss was low. In particular the data of example 1 show for a PVP concentration slightly larger than the minimum concentration calculated by the relation in claim 1 of application Serial No. 10/070,758 the reaction is still acceptable. The comparison of example 1 with example 2 and comparative example 1 shows that a further increase of the PVP concentration slightly improves iodine loss, while a decrease of the PVP concentration by the same amount dramatically deteriorates the reaction performance, since mixing is bad, insoluble sediments are formed, the reaction is slowed. Additionally, iodine sublimation in condenser occurs. The available iodine is dramatically decreased. Likewise, the comparison of example 1 and 3 with comparative example 2 shows that for constant PVP concentration a decrease of the K value below the lower limit require according to claim 1 of application Serial No. 10/070,758 leads to strong sublimation of iodine, which might lead to a clogging of the apparatus, and to a dramatic iodine loss. Moreover the available iodine is markedly below 9.5 % by weight. Thus, the data clearly demonstrate the significance of utilizing a PVP solution having a PVP concentration above the calculated minimum concentration calculated according to the relation in claim 1 of application Serial No. 10/070,758.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information or belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 101 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed at 67056 Ludwigshafen, this 29 day of May 2006.



(Signature of Declarant)

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